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Granular FePt–Ag thin films with uniform FePt particle size for high-density magnetic recording

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Abstract

The granular FePt–Ag thin films consisting of FePt particles dispersed in a Ag matrix were prepared by co-sputtering from FePt and Ag targets. The FePt particles in as-deposited film are soft magnetically disordered face-centered-cubic γ-FePt phase. The magnetic easy-direction of the as-deposited film is parallel to the film plane. The in-plane coercivities of the as-deposited films were below 700 Oe. The higher coercivity of the film will obtain after annealed treatment by transforming the γ-FePt phase into hard magnetically ordered face-centered-tetragonal (fct) γ_1-FePt phase. The in-plane coercivity of the annealed film was much larger than out-plane coercivity. The particle size of FePt in annealed film was increased with annealing temperature but decreased with increasing Ag content. The $(FePt)_{70}$–$(Ag)_{30}$ film with film thickness of 10 nm annealed at 600 °C for 15 min and ice–water quenched having an in-plane coercivity around 3.5 kOe. The M_s value of this film is about 350 emu cm^{-3} and the squareness is about 0.8. The FePt particle size in this film was very uniform and the FePt particles were isolated by Ag.

Keywords: Cosputtering; Granular FePt–Ag thin film; Magnetic properties

1. Introduction

For high density magnetic recording thin film, the magnetic grains must be small enough to be nanoparticle size and also requires that uniform size and isolated magnetic particles to reduce intergrain interaction, which leads to lower media noise [1–3]. However, small particle size will result in smaller K_uV/K_BT value (where K_u is the uniaxial magnetocrystalline anisotropy, V is the particle volume, K_B is the Bolzmann's constant and T is the absolute temperature), which leads increasing the thermal fluctuation of magnetization [4,5]. It well know that large K_u can resist thermal fluctuation of magnetization even the particle size is very small. The K_u value of FePt alloy is as high as 7×10^7 erg cm^{-3} and the saturation magnetization, M_s is about 680 emu cm^{-3} [6]. According to Stoner–Wohlfarth theory, for coherent rotation in a random distribution of noninteracting particles, the coercivity is $H_c = 0.96\ K_u/M_s$ [7], a very high H_c over 100 kOe of FePt alloy is expected.

Current studies have been focused on magnetically hard metal nanoparticle embedded in a nonmagnetic matrix for high density magnetic recording media. High in-plane coercivity materials consisting of the face-centered-tetragonal (fct) FePt equiatomic $L1_0$ phase dispersed in nonmagnetic matrices have been reported in FePt–C [8], FePt–SiO_2 [9], and FePt–Si_3N_4 [10] thin films. In this work, we investigate the magnetic properties and microstructure of granular FePt–Ag thin films which have not been studied before. We find that the granular $(FePt)_{70}$–$(Ag)_{30}$ thin film after annealing at 600 °C for 15 min had in-plane coercivity $H_{c//}$ of 3.5 kOe and the grain size of FePt is about 30 nm. It is suitable for applying in high-density magnetic recording media. The magnetic hardening mechanism of the $(FePt)_{70}$–$(Ag)_{30}$ thin film are also investigated.

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2. Experimental procedure

$(FePt)_{100-x}$–$(Ag)_x$ thin films ($x = 0$–50 vol.%) were deposited on (100) silicon wafer substrates at room temperature by co-sputtering high purity FePt (99.99%) and Ag (99.99%) targets with dc magnetron sputter. The substrabe was rotated at 75 rpm in order to attain uniform composition of the film. A 10 nm under layer of Ag and a 10 nm cover layer of Ag are used to protect the film from oxidation. The film thickness of magnetic layer is varied from 10 to 100 nm.

The chamber base pressure was 3×10^{-7} Torr and films were deposited under an argon pressure of 10 mTorr. The d.c. power of FePt was fixed at 40 W and the d.c. power of Ag is varied from 0 to 17 W. The deposition rate of the FePt was about 3.3 nm min^{-1}. The as-deposited film was sealed in quartz capsules and post-annealed in vacuum at various temperatures for 15 min, then was quenched in ice water.

The structures of the films were investigated by X-ray diffractometer (XRD) with Cu–K_α radiation. The film microstructure was observed by transmission electron microscopy (TEM). Thickness of the film was measured by an atomic force microscopy (AFM). Magnetic properties were measured by a vibrating sample magnetometer (VSM) and a superconducting quantum interference device (SQUID) at room temperature, with maximum applied fields were 13 and 50 kOe, respectively. The average grain size of the film was measured by the TEM bright field image. Composition and homogeneity of the film were determined by energy disperse spectrum (EDS) and the depth profiles of elements in the film were analyzed by Auger electron spectroscopy (AES).

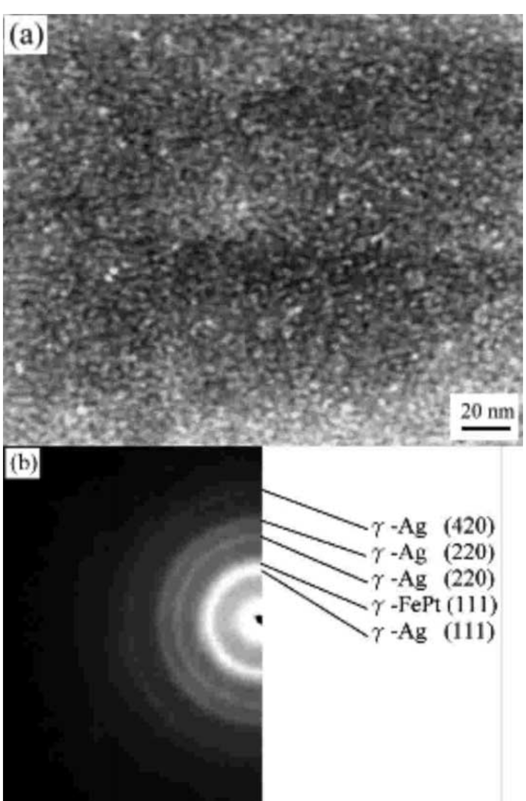

Fig. 1. TEM micrograph and electron diffraction pattern of the as-deposited $(FePt)_{70}$–$(Ag)_{30}$ film. (a) is the bright field image and (b) is the SAD pattern of (a).

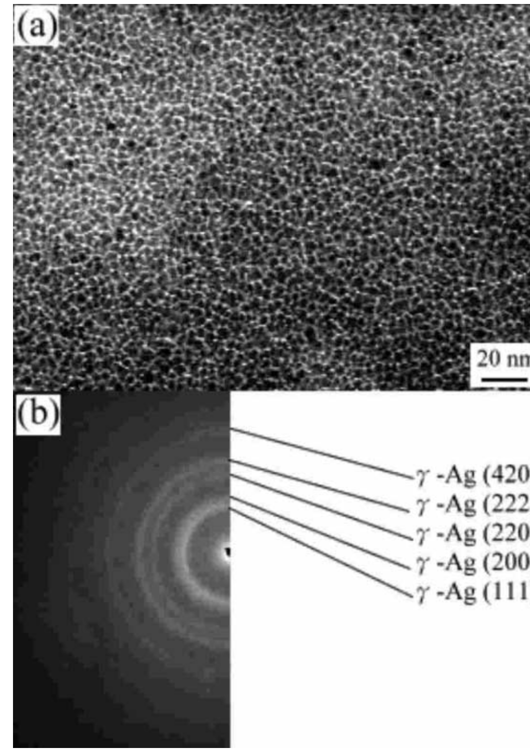

Fig. 2. TEM micrograph and electron diffraction pattern of the as-deposited pure Ag film, (a) is the bright field image, and (b) is the SAD pattern of (a). The film thickness is 10 nm.

3. Results and discussion

3.1. As-deposited FePt–Ag films

Fig. 1(a) is a TEM bright field image of the as-deposited $(FePt)_{70}$–$(Ag)_{30}$ film. The film thickness is 10 nm. It shows nanocrystalline structure inside the film and the average grain size of FePt (white grains) is about 3 nm. Comparing the areas of white and black grains of the TEM bright field image with the percentages of FePt and Ag within the film, it indicates that the white grain is FePt. Fig. 1(b) is the electron selected area diffraction pattern (SAD) of Fig. 1(a), it shows the structure of the as-deposited film contains not only fcc γ-FePt phase but also fcc γ-Ag phase. Fig. 2(a) and (b) are the TEM bright field image and SAD pattern of the as-deposited pure Ag film, respectively. We can see that the structure of the film is crystalline fcc γ-Ag phase and the grain size of the film is about 5 nm. Its grain color is black.

Fig. 3 shows the relationships among the saturation magnetization M_s, the in-plane coercivity $H_{c//}$, the out-plane coercivity $H_{c\perp}$ and the Ag content of various as-deposited $(FePt)_{100-x}$–$(Ag)_x$ films. We can see that M_s value of the pure FePt film is about 630 emu cm^{-3}, and it decreases linearly as the Ag content increases. As the Ag content is up to 50 vol.%, M_s value of the film is decreases to 310 emu cm^{-3} that is just the 1/2 of pure FePt. Since Ag is non-magnetic, increase of Ag will dilute the M_s value of the film. But $H_{c//}$ ascends slightly as the Ag content rises. $H_{c//}$ value for the pure FePt film is about 400 Oe, but it will rise to about 650 Oe as the Ag content increases to 30–50 vol.%. The increase of $H_{c//}$ may be due to that the increase of Ag makes the in-plane stress of the film become larger. However, $H_{c\perp}$ does not seem to be influenced by Ag content and keeps at about 55 Oe. Obviously, the magnetic easy-direction of the as-deposited films is parallel to the film plane. But, both $H_{c//}$ and $H_{c\perp}$ of the as-deposited film are not high enough for magnetic recording media application.

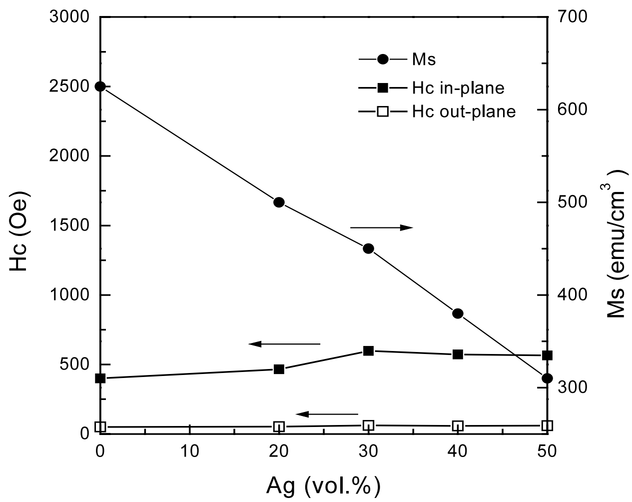

Fig. 3. The relationships among M_s, H_c, and Ag content of the as-deposited $(FePt)_{100-x}$–$(Ag)_x$ film. The film thickness is 10 nm.

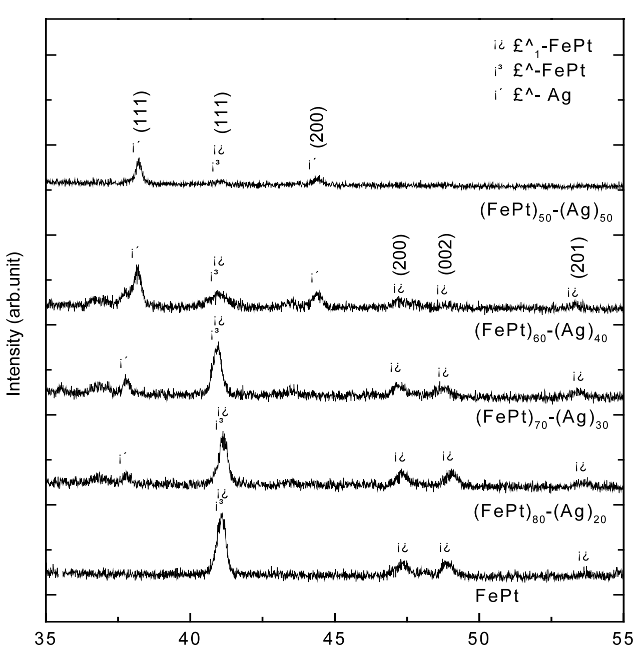

Fig. 4. XRD patterns of various $(FePt)_{100-x}$–$(Ag)_x$ thin films which annealed at 600 °C and the film thickness is 10 nm.

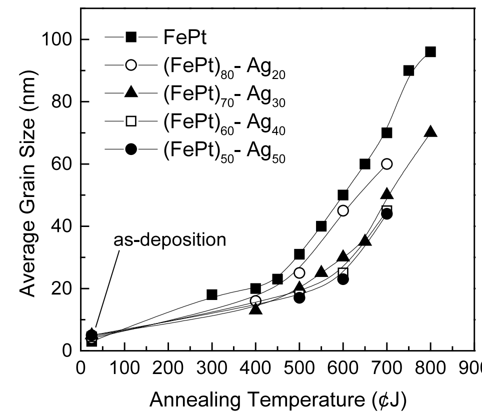

Fig. 5. Variations of average FePt grain size with annealing temperature for various annealed $(FePt)_{100-x}$–$(Ag)_x$ thin films. The film thickness is 10 nm.

3.2. Post-annealed FePt–Ag films

Fig. 4 is the XRD patterns of various annealed $(FePt)_{100-x}$–$(Ag)_x$ films. It shows that the intensity of the peaks of γ_1-FePt phase decreases as the Ag content increased. Since the thermal expansion coefficient of Ag is larger than that of FePt, the increase of Ag content will restrain the phase transformation of FePt from fcc γ-phase to fctγ_1-phase. And we can see that annealed pure Ag film still remains the fcc structure. The shift of the peaks at higher Ag content is due to the variation of the stress within the film.

Fig. 5 shows the relationships between annealing temperature and average grain size of FePt in various $(FePt)_{100-x}$–$(Ag)_x$ thin films. We find that the grain sizes of FePt of all the as-deposited films are smaller than 5 nm but they increase rapidly with the annealing temperature. At same annealing temperature, the FePt grain size is decreased as Ag content is increased. After annealing at 600 °C, the grain sizes of the pure FePt film, the $(FePt)_{70}$–$(Ag)_{30}$ film, and the $(FePt)_{50}$–$(Ag)_{50}$ film are 50, 30 and 20 nm, respectively. The surrounding Ag represses the growth of FePt grains in the film.

Fig. 6 shows the relationships among $H_{c//}$, $H_{c\perp}$ and the annealing temperature T_{an} of various $(FePt)_{100-x}$–$(Ag)_x$ films. $H_{c//}$ gently rises as T_{an} moves upward from

400 to 450 °C, but it climbs up promptly when T_{an} is higher than 450 °C. This is because the FePt transforms from the soft magnetic γ-phase to the hard magnetic γ_1-phase as $T_{an} > 450$ °C. Maximum $H_{c//}$ of all the films occurs at $T_{an} \approx 600$ °C. The maximum $H_{c//}$ is about 4.5 kOe for pure FePt but it will drop to about 3.5 kOe as Ag content increases to 30 vol.%. This is due to the increase of Ag content inhibits the growth of the FePt grains and makes the grains smaller and deviates from single-domain size of FePt. On the other hand, the XRD analysis reveals that the increase of Ag content would reduce the phase transformation of γ-FePt to γ_1-FePt phase. This can be another reason for its lower $H_{c//}$. When $T_{an} > 700$ °C, $H_{c//}$ drops very fast because the $(FePt)_{100-x}$–$(Ag)_x$ film takes reaction with the Si substrate. Fig. 7 shows the AES signal as a function of sputter time of the $(FePt)_{70}$–$(Ag)_{30}$ films with (a) as-deposited, (b) $T_{an} = 600$ °C and (c) $T_{an} = 750$ °C, respectively. The thickness of magnetic layer is 10 nm, and both the Ag under layer and cover layer are all 10 nm. It verifies Si atoms of the substrate has diffused into the magnetic layer as $T_{an} = 750$ °C, as shown in Fig. 7(c). This explains why $H_{c//}$ decreases abruptly about 750 °C as shown in Fig. 6. The maximum $H_{c\perp}$ for the as-deposited pure FePt film is about 2 kOe at $T_{an} = 600$ °C, but it will down to 1 kOe as Ag content increases to 30 vol.%, as show in Fig. 6. The relationships between $H_{c\perp}$ and T_{an} have the same tendency as that of $H_{c//}$. For any T_{an}, the $H_{c\perp}$ value is always smaller than $H_{c//}$, so the magnetic easy-direction of these annealed $(FePt)_{100-x}$–$(Ag)_x$ films is parallel to the film plane. From Fig. 7, we can also see that the Fe, Pt, and Ag elements distribute uniformly in the magnetic layer. For pure Ag film, the structure of the film still maintain crystalline fcc phase and the grain size is increased to about 25 nm after annealing at 600 °C, as shown in Fig. 8.

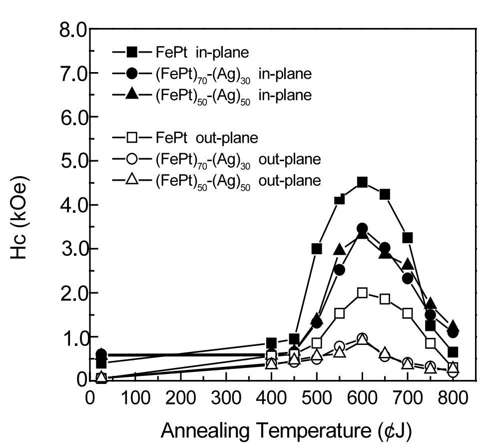

Fig. 6. Variations of $H_{c//}$ and $H_{c\perp}$with annealing temperature of various $(FePt)_{100-x}$–$(Ag)_x$ films; Ag contents of these films are 0, 30, and 50 vol.%, respectively. The film thickness is 10 nm.

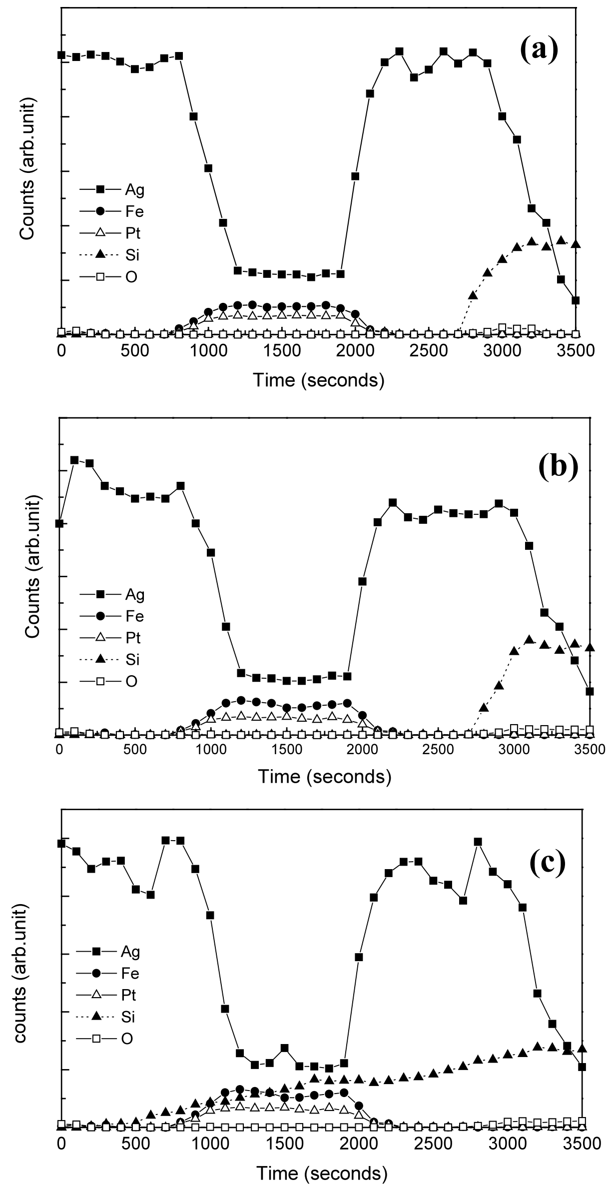

Fig. 7. AES depth profiles of the element as a function of sputter time for the $(FePt)_{70}$–$(Ag)_{30}$ thin films with (a) as-deposited, (b) $T_{an} =$ 600 °C, (c) $T_{an} = 750$ °C, respectively.

Fig. 9(a) is a TEM bright field image of the $(FePt)_{70}$–$(Ag)_{30}$ film which annealed at 600 °C. It shows that the magnetic FePt grains (white grains) are all surrounded by the non-magnetic Ag and well separated. This is responsible for the reduction in the interparticle interaction and thus can lower the noise of the recording medium [2,3]. The grain size of these FePt grains is uniform and it is about 30 nm. Shape of the FePt particles is like rice. Fig. 9(b) is the electron diffraction pattern of Fig. 9(a), it verifies the structure of the FePt phase have transformed from γ-FePt to γ_1-FePt, but Ag still remains the fcc structure as that of the as-deposited film. Fig. 10 shows the M–H loop of this film. We can see that its M_s value is about 350 emu cm^{-3}, $H_{c//}$ is about 3.5 kOe and the squareness (M_r/M_s) is about 0.8.

Fig. 11 shows the relationships among M_s, H_c, and Ag content in $(FePt)_{100-x}$–$(Ag)_x$ films which are annealed at 600 °C. Since the increase of non-magnetic Ag will dilute M_s value of the film, M_s value of the annealed FePt–Ag films decreases as the Ag content increases. M_s declines from 550 to 200 emu cm^{-3} when the Ag content is increased from 0 to 50 vol.%. The $H_{c//}$ value decreases from 4.6 to 3.5 kOe when the content of Ag is increased form 0 to 30 vol.%, but $H_{c//}$ value does not change distinctly when the Ag content is more than 30 vol.%. The $H_{c//}$ values are 3.5 and 3.3 kOe for 40 and 50 vol.% Ag, respectively. Since the increase of Ag content will restrain the growth of the FePt grains in the film and makes the grain size smaller and deviates from single-domain size, the $H_{c//}$ values decrease as Ag content is increased. However, the internal stress of the film and the interaction of the magnetic particles would also affect the $H_{c//}$ value. The change of $H_{c\perp}$ with Ag content corresponds to the tendency of $H_{c//}$, but $H_{c\perp}$ is always smaller than $H_{c//}$ no matter how much the Ag content is.

Fig. 12 illustrates the relationships between Hc values and film thickness of various $(FePt)_{100-x}$–$(Ag)_x$ films, which are annealed at 600 °C. It shows both $H_{c//}$ and $H_{c\perp}$ rise as the film thickness increases. Since thicker the film is, FePt grain size is larger and closer to the single domain size of the FePt grains. The grain growth will constrain by film thickness as grain size is larger than film thickness. For the pure FePt film, its $H_{c//}$ can reach 13 kOe when the film thickness is 75 nm. At this film thickness, the grain size of γ_1-FePt is just in the range of the single domain size, which is about 90 nm [6]. The $H_{c//}$ value decreases as film thickness higher than 75 nm. This phenomenon is consistence with the

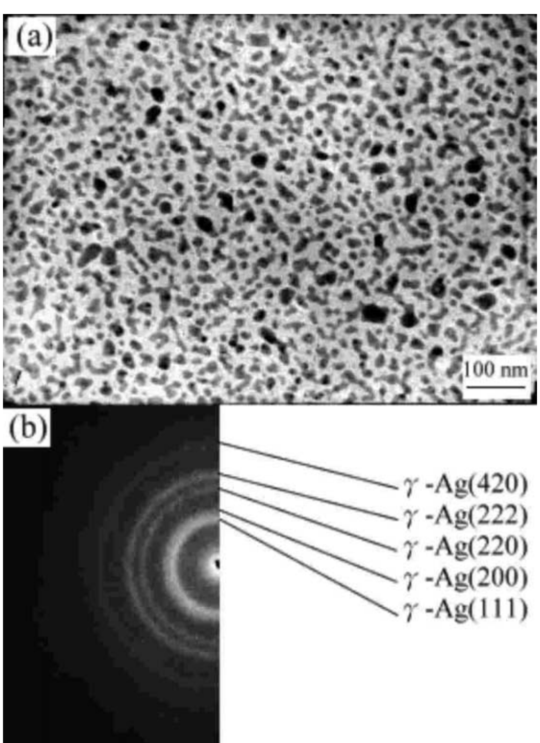

Fig. 8. (a) Is the TEM bright field image, and (b) is the SAD pattern of the pure Ag film which annealed at 600 °C for 15 min. The film thickness is 10 nm.

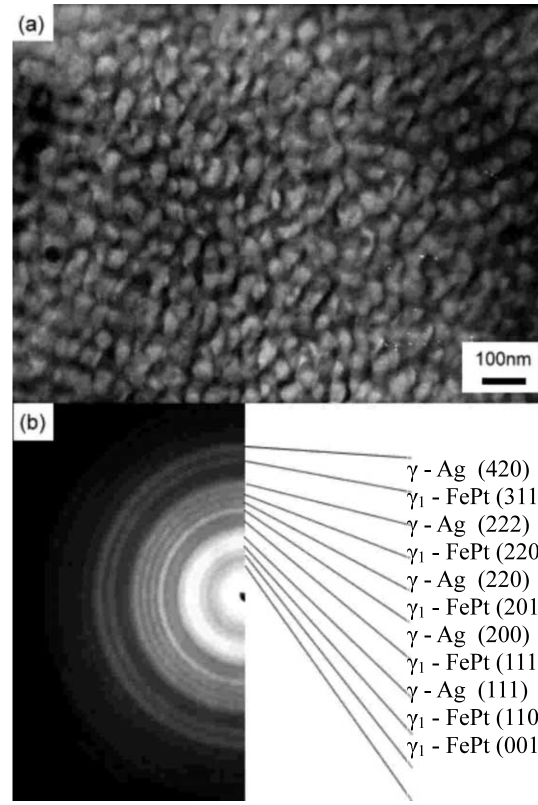

Fig. 9. (a) TEM bright field image of the $(FePt)_{70}$–$(Ag)_{30}$ thin film which is annealed at 600 °C. (b) is the SAD pattern of the film. The film thickness is 10 nm.

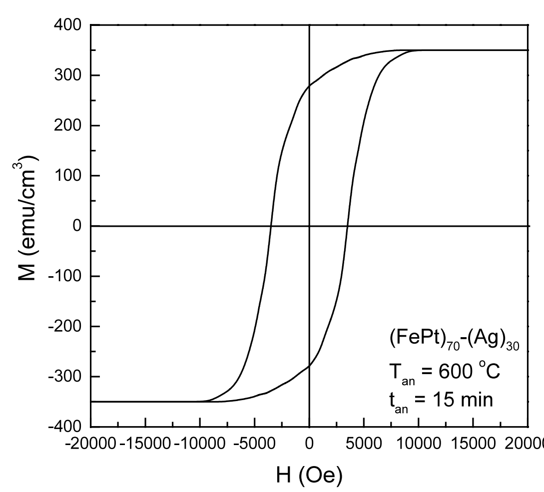

Fig. 10. The M–H loop of $(FePt)_{70}$–$(Ag)_{30}$ thin film which is annealed at 600 °C. Thickness of the film is 10 nm.

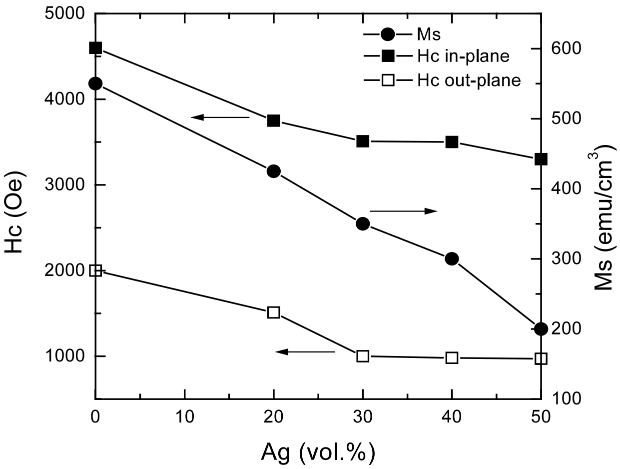

Fig. 11. The relationships among M_s, H_c, and Ag content of various annealed $(FePt)_{100-x}$–$(Ag)_x$ films. The annealing temperature is 600 °C and the film thickness is 10 nm.

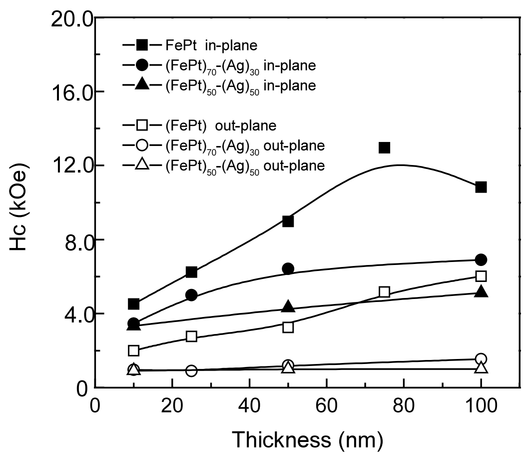

Fig. 12. Variation of H_c with film thickness of various annealed $(FePt)_{100-x}$–$(Ag)_x$ films. The film is annealed at 600 °C.

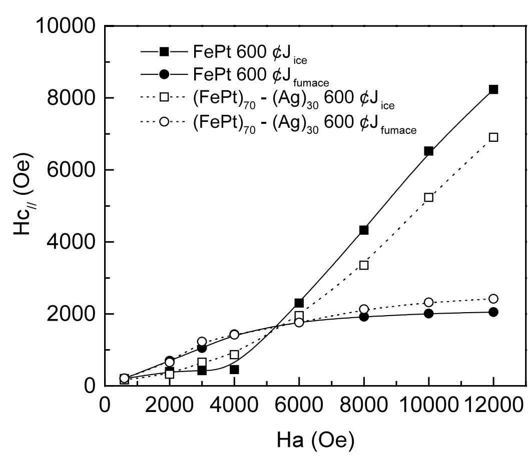

Fig. 13. $H_{c//}$ vs. H_a of various annealed FePt and $(FePt)_{70}$–$(Ag)_{30}$ thin films. The film thickness is 10 nm.

observation of Watanabe et al. [11] for pure FePt film. Both $H_{c//}$ and $H_{c\perp}$ drop as the Ag content increases whatever the film thickness is, because the increase of Ag content will repress the grain growth and makes the phase transform from γ-FePt to γ_1-FePt more difficult.

Fig. 13 shows variations of the applied field Ha with $H_{c//}$ values of various annealed $(FePt)_{100-x}$–$(Ag)_x$ films. The annealing temperature is 600 °C and the films are furnace cooling and ice water quench after annealing, respectively. The $H_{c//}$ values are obtained from the minor loops of the VSM measurement to examine the magnetic hardening mechanism of the film. We can see that the pure FePt film quenched with iced water has the domain wall pinning mechanism, but the film slowly cooled in a furnace (with the cooling rate of 4 °C min^{-1}) has the domain nucleation mechanism. It shows that the shape of the curve of the iced water quenched $(FePt)_{70}$–$(Ag)_{30}$ film is similar to that of iced water quenched pure FePt film, and the curve of the $(FePt)_{70}$–$(Ag)_{30}$ film cooled in a furnace is close to the pure FePt film cooled in a furnace. Distinctly, disperse FePt grains in Ag matrix does not change the magnetic hardening mechanism of the film. Excellent heat conductivity of Ag can be the reason to make the $(FePt)_{70}$–$(Ag)_{30}$ film dissipate heat as fast as that of the pure FePt film after heating. According to Tanaka et al. [12], anti-phase boundaries and various orientations of γ_1-FePt twins interfaces generate pinning sites and make the magnetic hardening mechanism of γ_1-FePt creates domain wall pinning mechanism. Consequently, addition of Ag in to FePt film has no significant influence upon the number of pinning sites.

4. Conclusions

Granular FePt–Ag thin films consisting of magnetic FePt particles embedded in non-magnetic Ag matrix were successfully prepared with controlled FePt particle size and magnetic properties of the film by varying film thickness, Ag content, and annealing temperature. The in-plane coercivity was much larger than the out-plane coercivity for as-deposited and annealed films. The particle size of FePt in annealed film was increased with annealing temperature but decreased with increasing Ag content. The $(FePt)_{70}$–$(Ag)_{30}$ film with film thickness of 10 nm annealed at 600 °C for 15 min and ice–water quenched having an in-plane coercivity around 3.5 kOe. The M_s value of this film is about 350 emu cm^{-3} and the squareness is about 0.8. TEM analysis exhibited that the FePt particles in this film were isolated by Ag and the FePt particle size was very uniform, it is about 30 nm. This film can be used for high density magnetic recording media with low media noise.

Acknowledgements

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References

[1] I. Kaitsu, A. Inomata, I. Okamoto, M. Shinohara, IEEE Trans. Magn. 31 (1998) 1591.
[2] R. Ranjan, J.A. Christner, D.P. Ravipati, IEEE Trans. Magn. 26 (1990) 322.
[3] Y.H. Lee, J.P. Wang, L. Lu, J. Appl. Phys. 87 (2000) 6346.
[4] J.I. Gittleman, B. Abeles, S. Bozowski, Phys. Rev. B9 (1974) 3891.
[5] T. Shimatsu, J.C. Lodder, Y. Sugita, Y. Nakamura, IEEE Trans. Magn. 35 (1999) 2697.
[6] C.M. Kuo, P.C. Kuo, H.C. Wu, J. Appl. Phys. 85 (1999) 2264.
[7] B.D. Cullity, Introduction to Magnetic Materials, Addison-Wesley, Reading, MA, 1972.
[8] J.A. Christodoulides, Y. Huang, Y. Zhang, G.C. Hadjipanayis, I. Panagiotopoulos, D. Niarchos, J. Appl. Phys. 87 (2000) 6938.
[9] C.P. Luo, S.H. Liou, D.J. Sellmyer, J. Appl. Phys. 87 (2000) 6941.
[10] C.M. Kuo, P.C. Kuo, J. Appl. Phys. 87 (2000) 419.
[11] M. Watanabe, T. Nakayama, K. Watanabe, T. Hirayama, A. Tonomura, Mater. Trans. JIM 37 (1996) 489.
[12] Y. Tanaka, N. Kimura, K. Hono, K. Yasuda, T. Sakurai, J. Magn. Magn. Mater. 170 (1997) 289.